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on

ADVANCED DIAGNOSTICS FOR REACTING FLOWS

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Prepared for

AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

For the Period

October 1, 1989 to September 30, 1990

Submitted by

R. K. Hanson, Principal Investigator

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Mechanical Engineering Department
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1.0 INTRODUCTION

Progress is reported for the past year of an interdisciplinary program aimed at establishing advanced optical diagnostic techniques applicable to combustion gases and plasmas, with some emphasis on high speed flows. The primary flowfield parameters of interest are species concentrations (including electrons), temperature, mass density, pressure, and velocity, and quantities derivable from these parameters such as mass flow rate (from the product of density and velocity). The techniques under study are based on laser spectroscopy, particularly laser absorption and laser-induced fluorescence, with the latter capable of providing both single-point and multi-point (2-d and 3-d) measurements. Laser sources include tunable cw lasers (ring dye and semiconductor diode lasers) and tunable pulsed lasers (excimer-pumped dye and narrow-linewidth excimer). The cw lasers are spectrally narrow, allowing study of a new class of techniques based on spectral lineshapes and shifts, while the pulsed lasers provide intense bursts of photons needed for techniques based on light-scattering phenomena. Accomplishments of note include: the first simultaneous measurements of temperature, pressure and velocity in high speed flows; the first application of tunable semiconductor diode lasers to absorption and fluorescence measurements in high temperature plasmas and supersonic flows; the first application of the planar laser-induced fluorescence (PLIF) technique to nonequilibrium shock tube flows; and further advances in the development of a laser-photolysis shock tube for rate constant measurements of elementary combustion reactions.

2.0 PROJECT SUMMARIES

Included in this section are summaries of progress in each of seven project areas. Additional descriptions of this work may be found in the publications listed in Section 3.2. Reprints of these papers are available on request. Personnel involved in these projects are listed in Section 4.0.

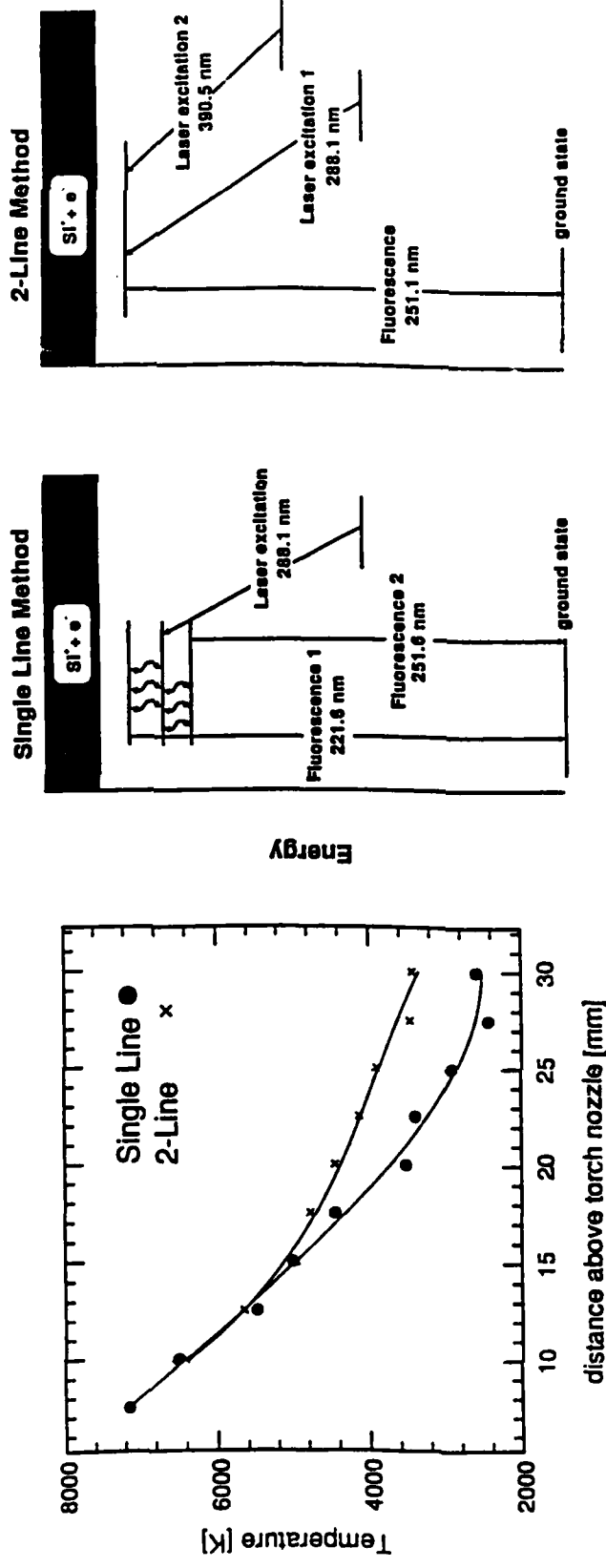
2.1 Plasma Diagnostics

During the past year we have conducted two series of tests in our atmospheric pressure, inductively-coupled RF plasma torch. The first series was aimed at evaluating single-point LIF strategies for measuring gas temperature using a tunable pulsed dye laser source; in this work the argon plasma was seeded with a low level of silane to generate trace levels of Si atoms. Two different strategies were explored, one based on single laser line excitation and the other based on two-line excitation. In the former case, a single transition of Si atoms is excited, and fluorescence is collected from two upper states, both populated through collisional transfer from the laser-coupled upper state. The ratio of these two LIF signals reflects the temperature in the upper states of the atom, which are closely coupled to the electron temperature of the plasma. The second method involves sequential laser-pumping of two lower levels of Si to a common upper level. The ratio of LIF signals in this case reflects the temperature in the lower levels of the atom, which is more likely to be near the kinetic temperature of the gaseous plasma. A graphic summary of this activity, indicating both the energy levels of Si utilized and sample temperature results, is shown in Fig. 1. Note particularly that measurements have been made up to 7000K and that generally good agreement is found between the two methods at high temperatures. The apparent discrepancy at low temperatures is probably a reflection of the fact that the plasma has "two temperatures," one describing the population distribution in the upper states of Si and the other reflecting the distribution in the lower states. This multiple-temperature behavior is characteristic of plasma systems and illustrates the additional challenges involved in developing quantitative diagnostic methods for plasmas relative to combustion gases.

The second series of tests this year was aimed at exploring the use of cw semiconductor diode lasers (GaAlAs) as light sources for absorption and fluorescence measurements of temperature and electron number density. These low-cost lasers are evolving rapidly owing to their numerous practical applications (compact disc players, supermarket scanners, etc.), but they also offer exciting prospects for use in scientific applications. With regard to spectroscopic diagnostics, these lasers can be viewed as economical, rugged and compact sources of low power, cw, tunable wavelength light with relatively narrow spectral linewidths. Thus they represent possible replacements for currently employed cw dye lasers. Unfortunately, diode lasers have primarily been

LIF Thermometry in Atmospheric Pressure Plasmas

- Separate temperature measurements of upper/lower energy levels



- Plasma produced with 1 kW RF-powered torch (Ar seeded with Si)
- Luminosity (to 7000 K) overcome with pulsed laser/gated detection
- Multi-point measurements possible using gated 2-d detector
- Potential for simultaneous determination of N_e and T

Hanson/AFOSR

Figure 1. Approaches and results for LIF thermometry in plasmas.

developed for use at near-infrared wavelengths (especially 1.3 and 1.5 microns), and only recently have lasers become available at wavelengths below 1 micron where most electronic transitions of interest in atoms and molecules are located. At the present time, diode lasers are available for use in selected wavelength bands down to about 750 nm. In our recent work we have utilized a diode laser which operates in the 802-815 nm range, which conveniently overlaps the strong 811.5 nm $4s^3P_2 \rightarrow 4p^3D_3$ transition of argon linking the first excited state of argon (4s) with the second excited state (4p).

The experimental arrangement utilized to study both absorption and fluorescence diagnostic strategies is shown in Fig. 2. The diode laser is rapidly swept in wavelength

Plasma Diagnostics using Tunable Diode Lasers

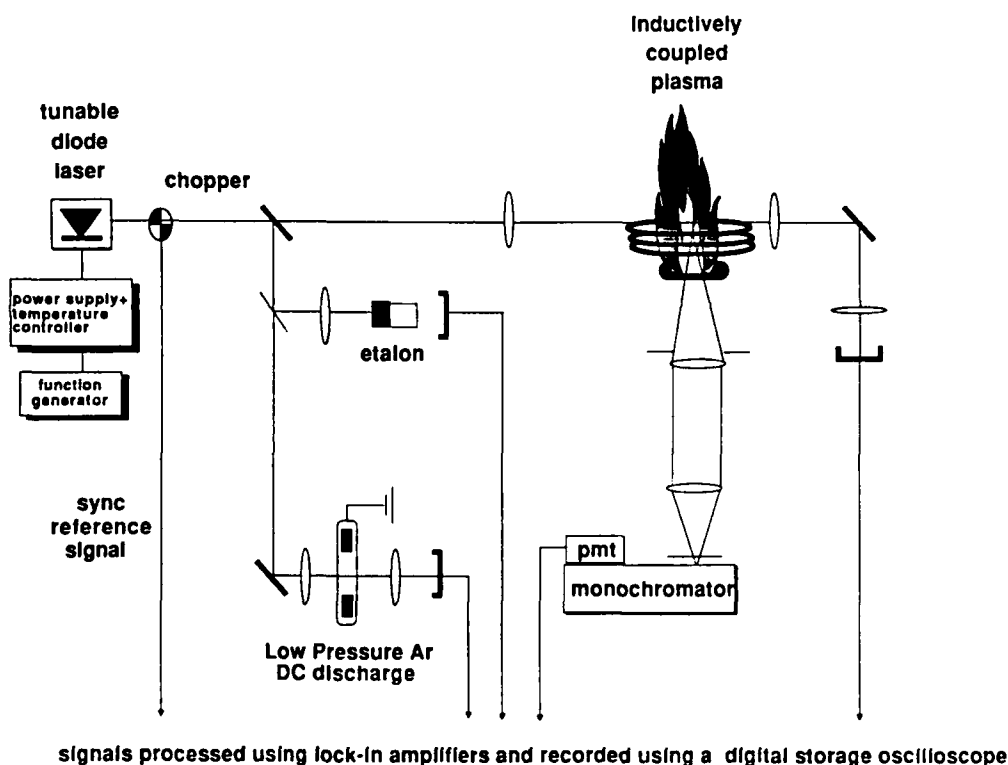


Figure 2. Laser Induced Fluorescence/Absorption diagnostics in an atmospheric pressure plasma using a tunable diode laser.

across the absorption transition of interest, providing a fully resolved record of the absorption lineshape. An example result, recorded at a height of 1.3 cm above the torch exit is shown in Fig. 3. The absorption data can be used to infer gas temperature (through Doppler broadening of the line) and electron density (through the Stark-induced shift in the line position). Since the lineshape also is influenced by Stark broadening, the linewidth data can be interpreted to yield a second, independent determination of the electron density. As

indicated on Fig. 3, excellent agreement is found between the two measurements of electron density; the inferred temperature is 6100 K, in agreement with the results reported above based on single-point LIF. Finally, it can be noted that the fractional absorption measured across the width of the plasma can be used to infer the population density in the absorbing excited state and hence a population temperature for this level.

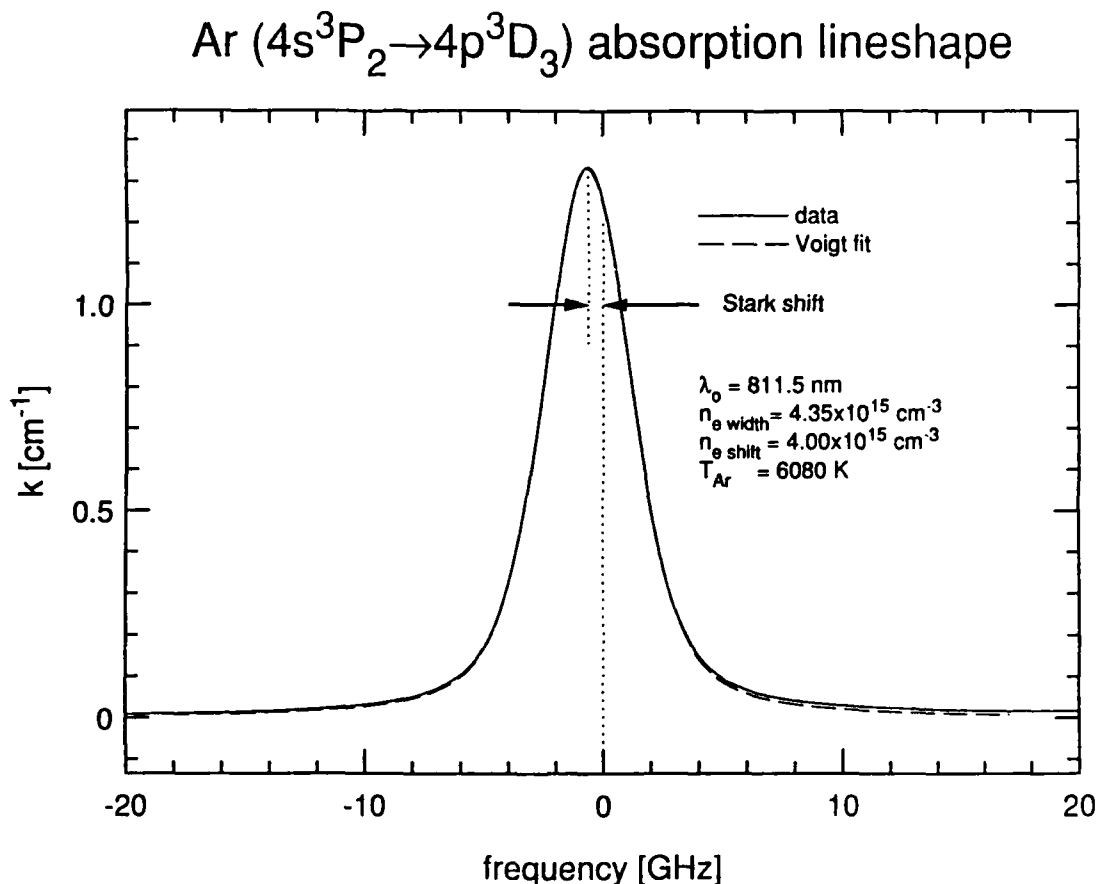


Figure 3. Absorption lineshape data for argon plasma; inferred temperature and electron density are indicated on figure.

Quite recently, we have extended this work by recording the absorption lineshape using fluorescence (collected at right angles to the laser beam). Although these scattered-light signals are significantly weaker, the LIF approach has two significant virtues: (1) the measurement is spatially resolved; and (2) the absolute signal level can be used to infer the electronic quenching rate, which will lead to useful information on the electron cross-section for collisional quenching.

These initial experiments apparently represent the first use of diode lasers in plasma diagnostic schemes. The success achieved in simultaneously determining multiple plasma properties using a single, economical laser source suggests that this is a promising line of research. We therefore plan to continue and to expand our research on this topic in the coming year.

2.2 Semiconductor Diode Laser Diagnostics for Gasdynamics Measurements

In addition to the work described above on plasma diagnostics, we have begun to explore the application of tunable GaAlAs diode lasers for absorption measurements of O_2 and H_2O in supersonic flows. The potential merits of these new laser sources are clear, and there is no doubt that such lasers will lead to new types of economical, rugged and compact instruments suitable for laboratory and, eventually, flight-based measurements. Our research during the past year has been aimed at gaining familiarity with these lasers and their capabilities for various wavelength modulation measurement strategies. A critical part of the effort has been to learn the essential aspects of the relevant absorption bands of both oxygen and water, the latter being particularly complex, and to generate the needed computer codes to predict temperature and pressure dependence of the absorption coefficients for selected transitions.

The experimental approach is summarized in Fig. 4. A commercial diode laser, operable near 760 nm, is rapidly scanned in wavelength (by modulating the injection current to the diode laser) over an interval encompassing one or more oxygen transitions in the A-band (a weak but well-documented band linking the ground electronic state and the metastable B-state of O_2). A calculated transmission spectrum for the R-branch of the band, assuming a 5 meter path through room air, is shown in Fig. 5. A virtue of the laser is that it can be modulated at very high frequencies, and in fact the scanning waveform also incorporates a modulation component, at frequency f , which allows sensitive detection of absorption at various harmonic frequencies, usually the second harmonic, $2f$. The $2f$ output for absorption measurements of the RR(17,17) and RQ(15,16) line pair (at 760.1 nm) in room temperature air is shown in Fig. 6. Note the good agreement between measured and computed $2f$ spectra, and also the fact that the measurements are completed in a few milliseconds. These data, acquired with a static gas sample, can be used to infer temperature (from the ratio of the two primary peaks) and the partial pressure of O_2 (from the magnitude of either peak). In the case of moving gas samples, the velocity (along the direction of the laser beam) can be inferred from the magnitude of the Doppler shift of the peaks.

Although absorption measurements represent a path-averaged result, there are many applications where such data are preferred over spatially resolved measurements. Combustion-related examples might include: mass flux measurements in an engine inlet, where the path integral of the product of oxygen density and velocity can be used to provide a measure of the total rate of air inflow to the engine; or combustor exit plane measurements of the path-averaged flux of combustion products (e.g., H_2O) which could be used in a control system requiring combustion efficiency information. Analogous measurements would be of great use in ground-based test facilities for aerodynamics and propulsion research, where on-line data for velocity, temperature, species concentration or mass flux, are needed. Certainly,

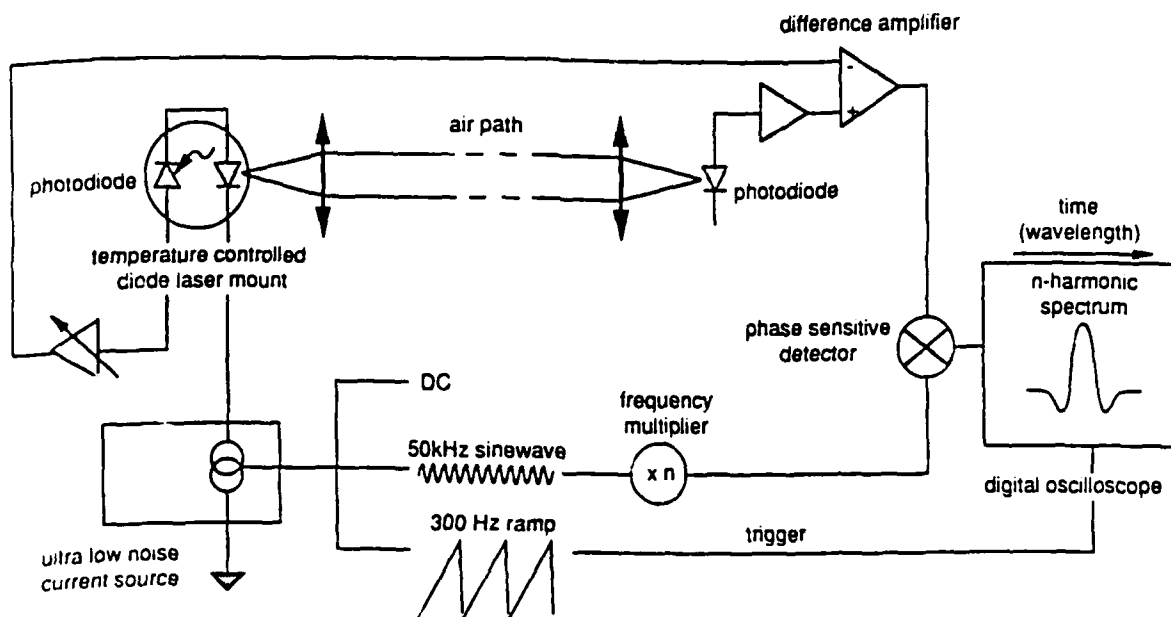


Figure 4. Experimental arrangement for tunable diode laser absorption.

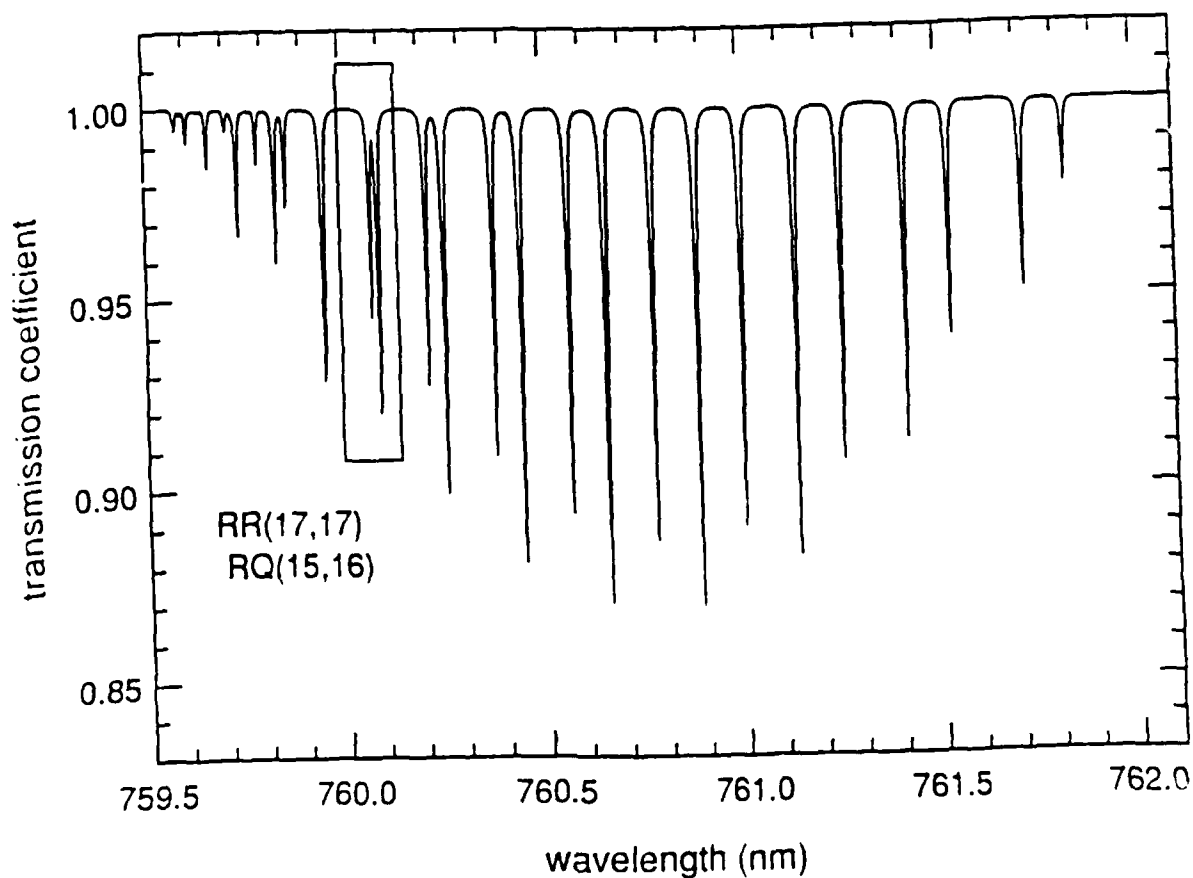


Figure 5. Calculated transmission for 5 meters of room air; R-branch of A-band.

instrumentation of the type indicated here is currently unavailable and would find considerable use in combustion/propulsion research, particularly since this instrumentation has the potential for being implemented in user-friendly packages once the optimum measurement strategies have been established.

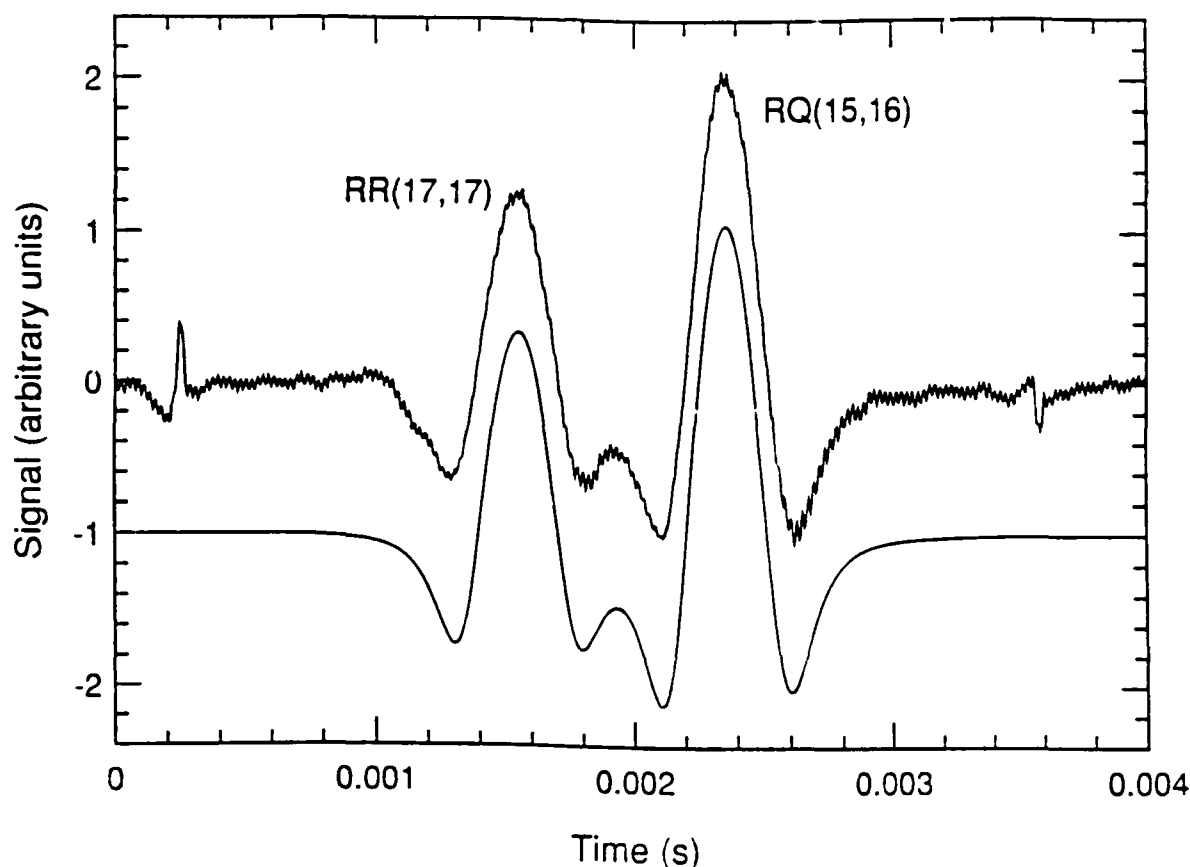


Figure 6. Measured and calculated second-harmonic absorption profiles for 5-m path of air at room temperature.

Our current research involves extension of the system shown in Fig. 4 to supersonic flows. In this case the modulation frequency has been increased to 1 MHz (with $2f$ at 2 MHz), and the scanning frequency to 10 kHz, so that measurements can be made rapidly (a scan is completed each 100 microseconds) as needed in pulsed flow facilities. The research is being carried out in a shock tube, which provides a convenient source of high-speed, high temperature air for about 1 millisecond. The challenge of the work is to establish modulation and detection strategies which are suitable for small levels of absorption, since the absorption features are quite weak. At present we are able to infer velocity to within about 10%, at a 10 kHz rate, in atmospheric pressure flows of air at velocities of about 1 km/sec with a pathlength of 20 cm. Measurements at lower repetition rates can be made with higher accuracy.

2.3 CW Ring Dye Laser Techniques

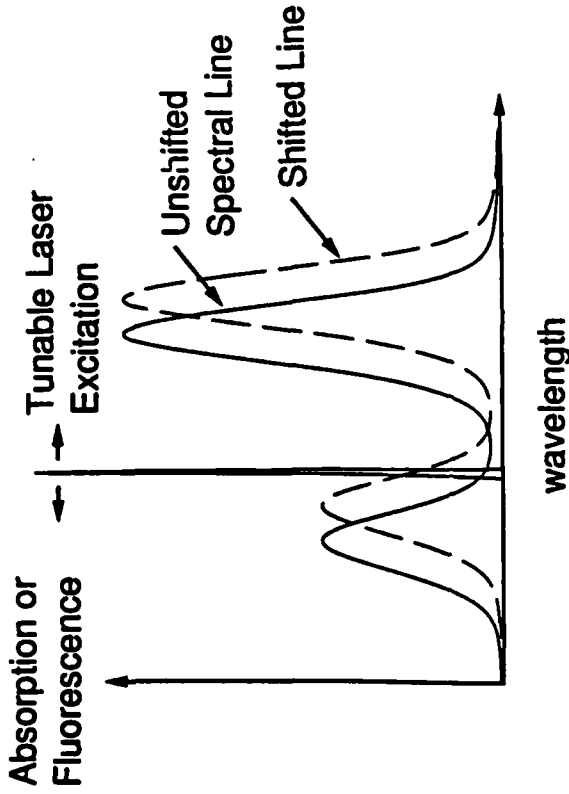
Continuous-wave (cw) laser sources offer certain advantages for laser diagnostics which have not yet been widely exploited. In our laboratory, for example, we've developed several diagnostics over the past 8 years based on cw single-mode ring dye lasers. Such lasers provide a continuous, low power source of nearly monochromatic light well suited for probing the details of spectral lineshapes using either absorption (line-of-sight) or single-point fluorescence (spatially resolved) measurements. The ability to fully resolve absorption lineshapes leads to several important measurements capabilities: (1) highly sensitive and quantitative species concentration measurements from the magnitude of the absorption (or fluorescence) signal; (2) temperature from the ratio of signals from different absorption lines; (3) pressure (or density) from the shape (width) of lines; (4) velocity from the Doppler shift of lines relative the spectral position of lines in a static sample; and (5) various derived quantities, such as mass flux which is given by the product of mass density and velocity. A schematic of this general measurement strategy is given in Fig. 7. It is important to note here that these various quantities can usually be obtained simultaneously, and, since these measurements are made by rapidly scanning in wavelength, the measurement repetition rate can be very high. In fact, some measurement strategies involve fixing the laser wavelength and probing the flow simultaneously from multiple directions, and in this case the measurements are continuous. It should be clear from the above discussion that measurement techniques based on cw lasers offer important capabilities not possible with pulsed laser techniques. The price paid, of course, is that cw lasers are low in power and hence are not generally suitable for imaging measurements which yield data for a large number of measurement points simultaneously. During the past year, there have been three accomplishments of note, discussed below.

Relatively early in the year, we set up an experiment to monitor OH in a bench-top supersonic combustor. The goal was to establish a method for simultaneously monitoring temperature, pressure and velocity at a single point in the flow using fluorescence monitoring of absorption probed with our wavelength modulation technique. We used the $R_1(7)/R_1(11)$ line pair of OH, which have a fortunate near coincidence in wavelength at 306.5 nm, and modulated our cw ring dye laser at about 3.5 kHz over the 2 cm^{-1} interval encompassing these lines. The LIF signal from a point (about $0.2\text{ mm} \times 0.2\text{ mm} \times 0.2\text{ mm}$ in extent) on the axis of the slightly supersonic flow exiting from a high-pressure methane-air combustor was monitored on a PMT, recorded on a digital scope, and post-processed to infer the desired quantities. The focussed laser beam from the argon ion-pumped ring dye laser was incident at 45 degrees to the flow axis to provide a Doppler shift in the LIF signal. An example result for a single scan of the laser (requires about 300 microseconds overall) is shown in Fig. 8.

Simultaneous Multi-Parameter Measurement Technique for Supersonic Flows

- Multiple Parameters Required to Characterize Compressible Flows

Temperature:	Intensity Ratio
Velocity:	Doppler Shift
Pressure:	Linewidth
Density:	$\rho = P / R T$
Mass Flux:	$\dot{m} = \rho V$



- Non-Intrusive Method Based on CW Laser Absorption or Fluorescence
- Two Configurations: LIF \rightarrow Spatially Resolved Data
Absorption \rightarrow Line-of-Sight Data
- Novel Rapid-Scanning Laser Enables 4 kHz Measurement Repetition Rate
- Accessible Species: OH, NO, O₂
- Possible Extensions to Higher Repetition Rates
- First High-Speed Multi-Parameter Measurement Technique

Figure 7. Laser wavelength modulation strategy for simultaneous multi-parameter measurements using absorption or fluorescence spectroscopy.

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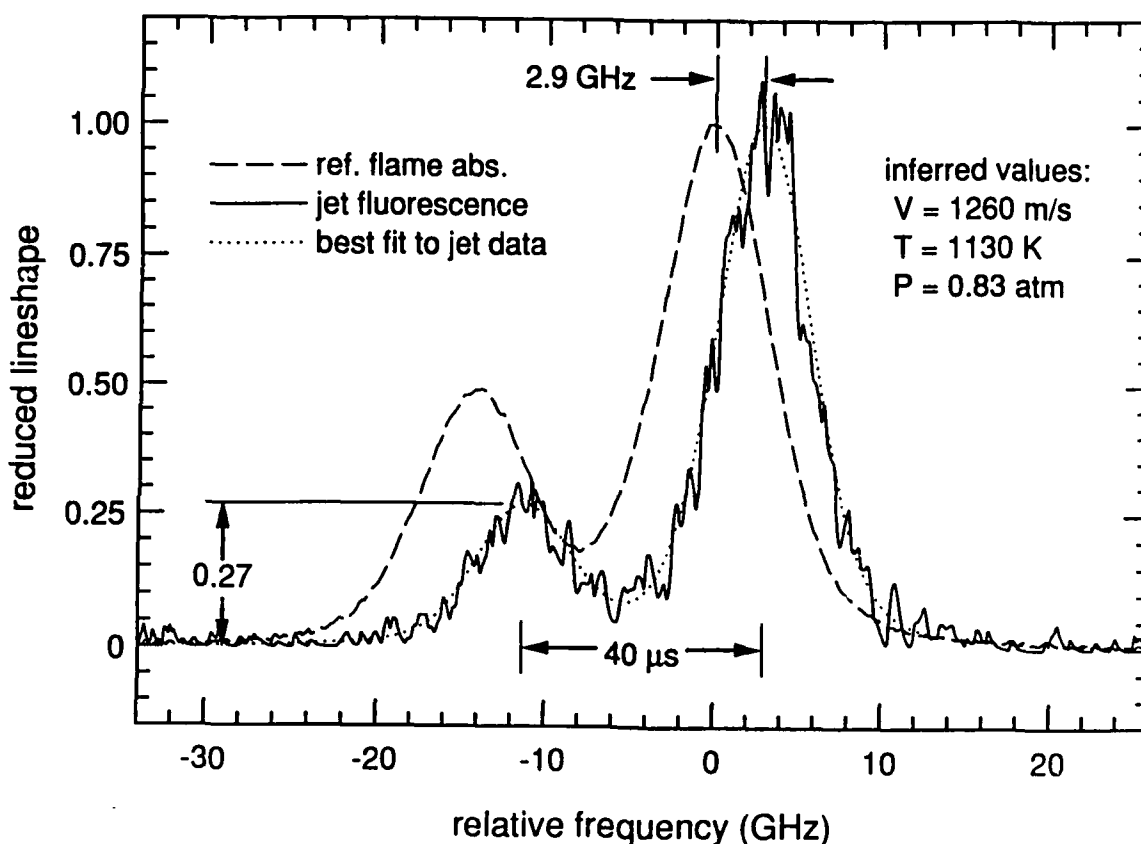


Figure 8. Single-point LIF data in OH for simultaneous measurements of velocity, temperature and pressure in supersonic combustion flow.

Note that the signal from a reference flame (essentially static flow) was recorded simultaneously to allow direct determination of the Doppler shift. As indicated on the figure, the inferred axial velocity is 1260 m/s, the temperature is 1130 K, and the pressure is 0.83 atm, all in good agreement with flowfield calculations. We believe that these are first results demonstrating the ability for simultaneous optical measurements of multiple flowfield properties in a reacting or nonreacting flow.

Our second accomplishment involved the successful application of wavelength modulation absorption spectroscopy in shock tube flows. This work was aimed at exploring concepts for line-of-sight-averaged quantities including velocity, temperature, density (or) pressure, and mass flux using the same general lineshape/shift/strength ideas employed above for OH LIF. The shock tube provides a convenient means of generating a very wide range of flow parameters under well-known conditions, and at the same time simulates some of the difficulties associated with measurements in high enthalpy, pulsed flow facilities. A critical objective in this research was the extension of the measurements to relevant new species, specifically NO and O₂ which are of course often present in combustion and other high temperature flows. This work entailed extending the capabilities of our ring dye laser to shorter wavelengths (215-230 nm), through use of a custom-built BBO frequency doubling crystal. The result is the first, to our knowledge, cw ring dye laser with this capability.

As an example of the various shock tube measurements completed, Fig. 9 provides a schematic diagram of the setup employed in experiments with NO using the $A \rightarrow X(0,0)$ band near 225 nm. Here a crossed-beam arrangement was used with absorption data

Ring Dye Laser Absorption in a Shock Tube

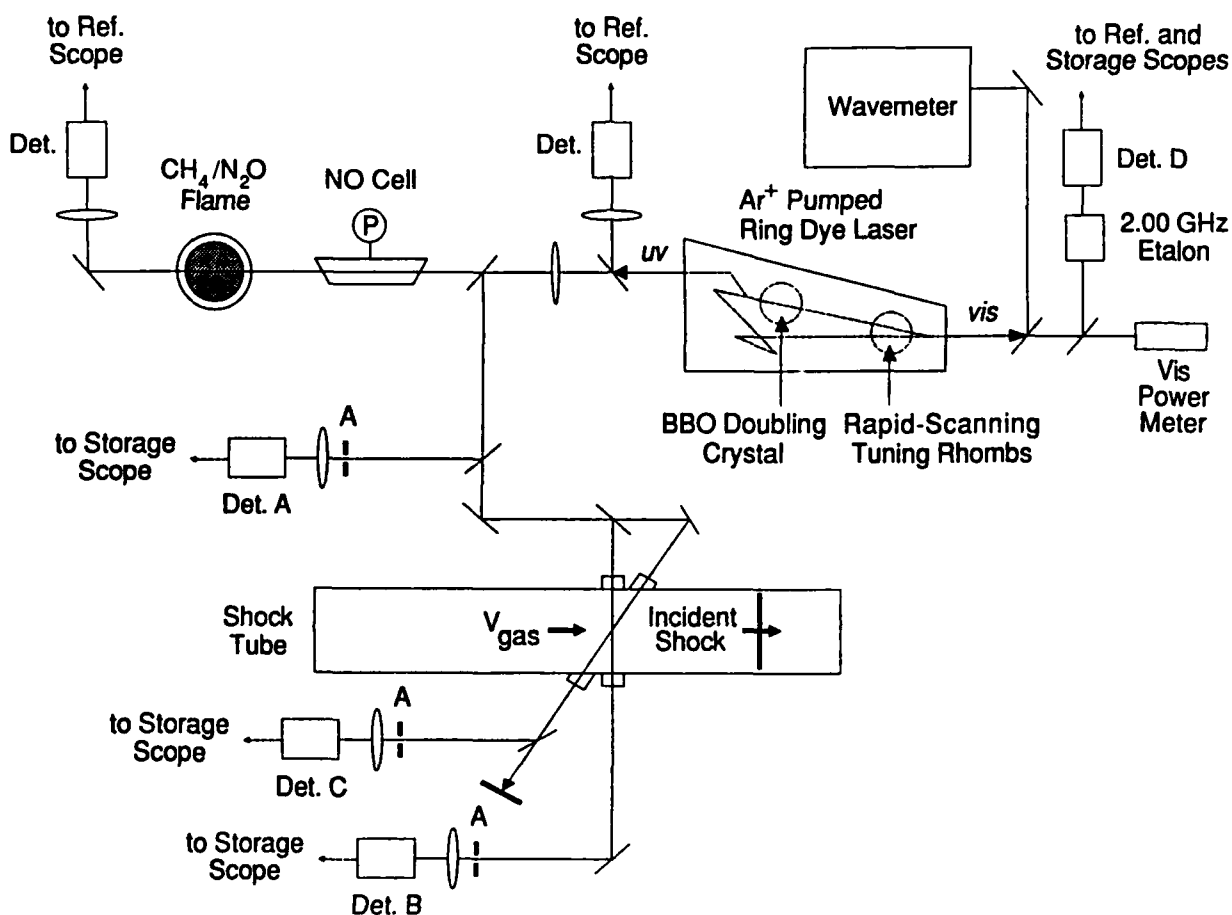


Figure 9. Experimental arrangement for wavelength modulation diagnostic in shock tube flow.

recorded along beams which cross the shock tube flow at two angles. As with OH, the laser wavelength is rapidly modulated (about 4 kHz repetition rate) to allow recording of two closely spaced NO transitions, thereby enabling inference of temperature from the ratio absorption in the two lines. The absolute strength of the absorption signal yields the path-averaged concentration (or, equivalently, the pressure since temperature is also determined), while the Doppler shift of the lines between the two beam directions yields the post-shock velocity of the gas, both at the laser repetition rate of 4 kHz. Excellent agreement was found between the measurements based on this detection strategy and the flowfield parameters computed using standard shock wave relations. As an example, measured and computed

velocities are compared in Fig. 10 for a range of velocity up to 1400 m/s. Similar measurements have been carried out for O_2 using the $B \rightarrow X$ Schumann-Runge system. In summary, these experiments on NO and O_2 provide important first demonstrations of the potential of laser diagnostics based on cw laser wavelength modulation concepts. Further work on this topic is planned for the coming year.

Absorption Velocimetry of NO

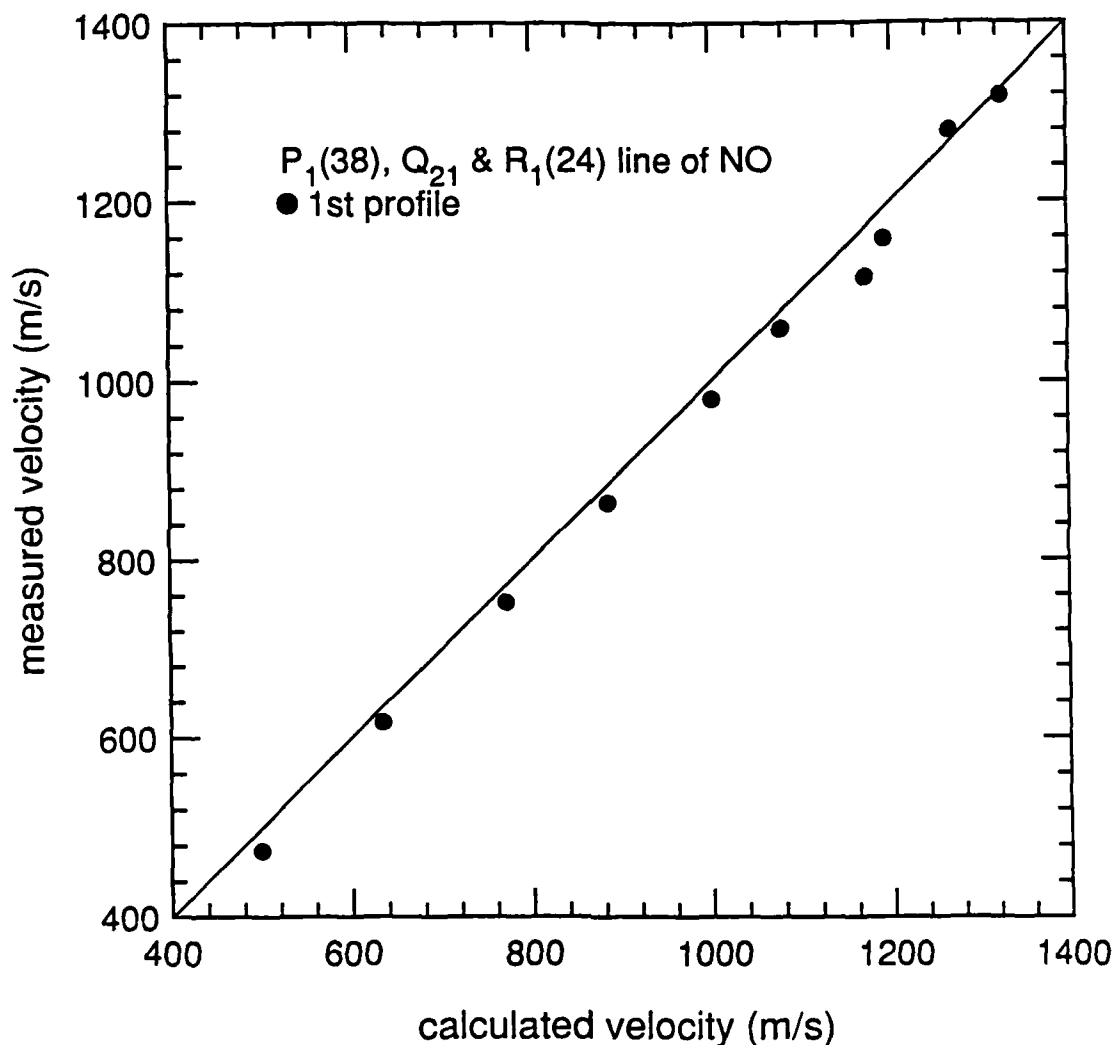


Figure 10. Velocity results obtained using wavelength modulation absorption spectroscopy of NO in a shock tube.

Finally, a third highlight for the past year was the demonstration of a simple, fixed-frequency scheme for continuous monitoring of velocity using a cw ring dye laser. The usual setup for these experiments was the same as shown in Fig. 9 except the laser wavelength was fixed at a spectral position off resonance for the absorption line (in either OH or NO). In this case, a simple ratio of the absorption along the two beam paths in the shock tube, normalized

by the relative beam lengths, yields the relative absorption lineshape functions for the gas probed. These functions are identical except for the Doppler shift associated with the angled beam, and hence this normalized signal can be used to infer the velocity as a continuous function of time behind the shock wave. The results are found to be in good agreement with calculated velocities. We are unaware of other current nonintrusive optical methods which yield velocity continuously. Future work to extend this approach to spatially resolved measurements through LIF detection is planned, at which point, if successful, the method would provide both spatial and temporal resolution in continuous velocity measurements.

In addition to the projects described above, we have maintained contact during the past year with the shock tunnel group at NASA Ames which has been implementing a wavelength modulation system to monitor OH concentration and temperature in its 16-inch tunnel. This is a collaborative effort which should lead to the transfer of our previously developed diagnostics technology into a hypersonic flow facility of some importance in current research on the National Aerospace Plane.

2.4 PLIF Imaging in Nonequilibrium Shock Tube Flows

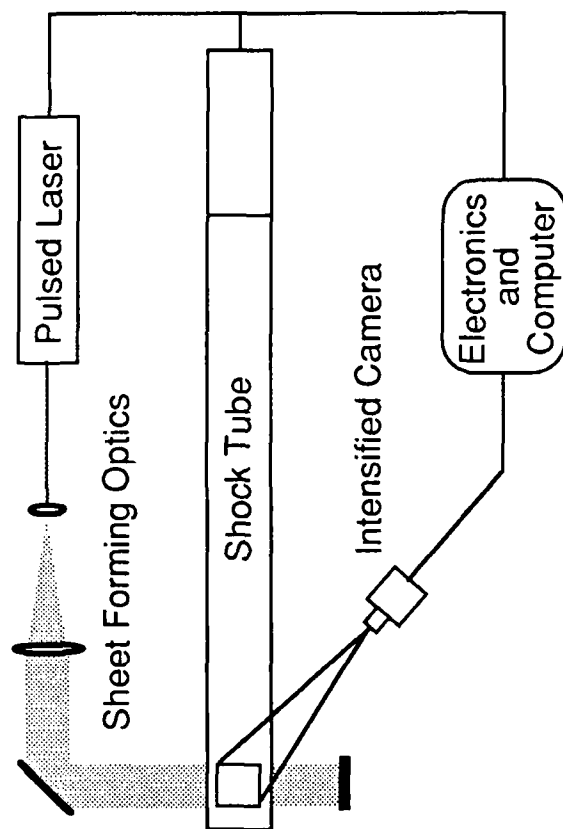
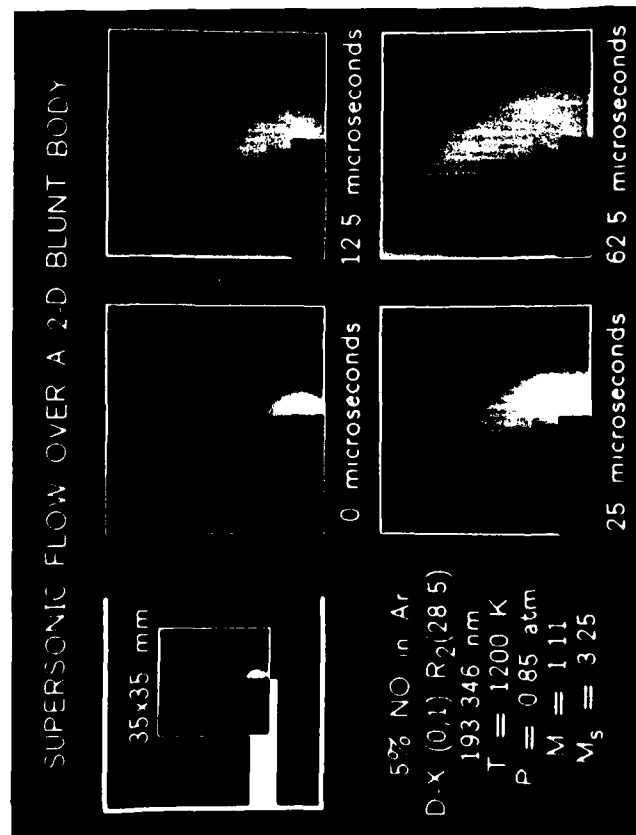
Nonequilibrium supersonic/hypersonic flows, relevant to current research on scramjets, pose new measurement problems for experimentalists. For example, experiments are often conducted in pulsed flow facilities in which the available measurement time is quite limited, thereby putting a premium on the ability to acquire complete data sets in very short times. In addition, many flows of interest exhibit a high degree of nonequilibrium, requiring experimental methods sensitive to such nonequilibrium effects. PLIF has high potential for dealing with both of these critical problems, in that the data provide population densities in specific quantum states of the species probed at a very large number of flowfield locations. During the past three years we have initiated research which addresses some of the problems inherent in extending PLIF to transient supersonic flows, and these experiments are now beginning to yield important results.

Most of the experiments have been conducted in a standard pressure-driven shock tube, built specifically for this work two years ago, and have emphasized measurements of either OH or NO. These species are attractive owing to their common presence in combustion and high temperature air flows of interest. Initial research was concerned with solving problems of synchronization of the shock wave position, the laser pulse and intensified camera gating. This can now be done with an uncertainty of less than one microsecond, or equivalently a few millimeters of shock location. More recently, the research has proceeded in two parallel paths, one involving imaging of OH in combusting flowfields and the other involving imaging of NO in flows with vibrational nonequilibrium.

Imaging of NO began with simple planar shock waves (incident and reflected shock waves) in NO-Ar and NO-N₂ mixtures. These experiments were ideal for investigating questions of spatial resolution in PLIF images in that the shock waves are known to be planar and very thin. We learned in these experiments that the fluorescence collection optics must be focussed with extreme care to avoid degradation of spatial resolution in the images; in addition, we found that commercial lenses, of high quality, produced much sharper images than simple lenses. These same normal shock wave experiments also served as a useful introduction to the study of vibrationally nonequilibrated flows, since the vibrational relaxation of NO can be readily computed for 1-d flows using known relaxation times. The quantity measured in the PLIF image is essentially the population density in a specific v'' , J'' state, which may be thought of as the product of two Boltzmann fractions, one for the vibrational mode and one for translation/rotation. Thus, by imaging (in successive shock wave experiments) states with different v'' (usually 0 and 1), the PLIF data can be used to generate a map of the vibrational temperature behind the shock wave. We found generally good agreement between these nonequilibrium temperatures and those computed using known relaxation time data. Most recently, we have been working to extend PLIF imaging in nonequilibrium flows which are either two or three dimensional. An example (see Fig. 11) is supersonic flow over a 2-d blunt body. The body is mounted on the shock tube end wall and protrudes upstream to the location of the PLIF observation window. An incident shock wave ($M = 3.25$) in a 5% NO-Ar mixture generates post-shock conditions of 1200 K and 0.85 atm which flow over the body at supersonic speeds. Images acquired at different instants of time clearly show the time-dependent formation of a bow shock with a nonequilibrium vibrational flowfield. Although in this case the imaging was done with a single laser (tunable 193 nm ArF) and camera, experiments involving excitation with two laser lines and cameras could be performed to give instantaneous vibrational temperatures and NO density on a single-shot basis. Other aspects of current work with NO involves selection of optimum laser wavelengths for excitation using both tunable 193 nm ArF excimer lasers (access to D-X system) and excimer-pumped dye lasers, the latter providing access to several vibrational bands in the A-X system.

PLIF imaging of OH has been carried out in two flowfields: (1) shock-induced ignition of H₂-O₂-Ar mixtures using a nonplanar end wall to generate flow nonuniformities; and (2) combustion of H₂ injected transversely into a supersonic air stream generated by an incident shock wave. The former experiments (see Ref. 18, listed in Sec. 3.2) utilized a grooved end wall to provide a nonuniform reflected shock flowfield which led to localized ignition of the combustible mixture. The strong 3-d nature of these flows, captured by the PLIF images, provided graphic evidence of the utility of PLIF for studying multidimensional flows. More recently, we have begun study of transverse injection of fuel into supersonic air streams. In these experiments, a pulsed jet of H₂ (from a 2 mm dia orifice in the shock tube side wall) is

PLIF Imaging of Hypersonic Flows in Vibrational Nonequilibrium



- First application of PLIF imaging to an impulse flow facility (shock tube)
- Spatially and temporally resolved, quantum state-specific measurements of NO
- Excitation of $v''=0$ and $v''=1$ levels of NO demonstrated with a tunable ArF laser
- Extension to a 2-laser/2-camera technique yields simultaneous T_{vib} and species concentration

Hanson/Stanford

Fig. 11. PLIF imaging in a shock tube.

activated just prior to passage of a strong incident shock wave in air. PLIF images are acquired at different times (following shock passage) and for different orientations of the illumination laser sheet. These images have revealed a surprising degree of structure in the burning interface between fuel and oxidizer, structure which is not apparent in frame-averaged data. Although our primary goal in this program is diagnostics research, rather than study of supersonic combustion, we believe that these initial images confirm the presence of large-scale organized structures in the combustion of transverse fuel jets in high-speed flows. These observations suggest the timeliness of a follow-on study, employing PLIF as a tool, to develop improved models of combusting jets in supersonic flows. Based on current data, it appears that a proper flowfield model should recognize the role of organized structures and, therefore, should not be based on conventional time-averaged jet-mixing concepts.

To our knowledge, the work at Stanford represents the first application of PLIF to hypersonic flows, flows with vibrational nonequilibrium, and to impulse flow facilities. The utility of PLIF for such flows is clear, and the results have already generated interest in the supersonic/hypersonic flow community.

2.5 Laser-Photolysis Shock Tube

The laser-photolysis shock tube is a new concept for generating controlled levels of free radical species in a high temperature environment. In brief, a shock tube, operated in the reflected shock mode, is used to heat a gas sample to desired high temperature conditions, and a pulsed excimer laser is used to illuminate portions of the gas with high intensity ultraviolet radiation. The UV photons act to photolyze a fraction of the initial molecules and thereby create a controlled level of free radicals. Such a clean, instantaneous source of combustion radicals is of value to propulsion science because it enables more direct studies of spectroscopic and reaction kinetic parameters of these species than have been possible in the past. The concept of a laser-photolysis shock tube originated in our laboratory, and the facility we've assembled is the first of its type. The facility has been operational for nearly three years and is being used in studies to develop absorption-based diagnostics for radical species and in kinetic studies of important combustion reactions.

During the past year our major accomplishment has been to extend the capabilities of the laser-photolysis shock tube to C- and N-atom reactions. This has included the establishment of suitable photolysis precursors for these atoms as well as the development of sensitive diagnostic schemes for monitoring atom concentration time histories. For N-atoms, we've found that pulsed ArF radiation (193 nm) of NO leads to instantaneous production of N and O. These N-atoms can then react with other molecules present in the mixture; a recent example has been the measurement of $N + H_2 \rightarrow NH + H$ in shock-heated NO-H₂-Ar mixtures. The reaction $N + NO \rightarrow N_2 + O$ has also been measured using NO-Ar mixtures

alone (only a small fraction of the NO undergoes photolysis). For C-atom kinetics, we've found that carbon suboxide (C_3O_2) is an attractive precursor. Illumination with pulsed 193 nm radiation yields (primarily) $C + 2 CO$. For cases in which the CO is relatively inert, reactions of C-atoms with other species (inert to the photolysis process) can be studied; recent examples are $C + H_2 \rightarrow CH + H$ and $C + O_2 \rightarrow CO + O$. The detection of the atom time histories, which leads to the rate coefficients of interest, is performed with atomic resonance absorption spectroscopy (ARAS). The resonance radiation, in the vacuum UV, is generated with microwave excitation of low-pressure N_2 -He and CO-He mixtures for N-atoms (119.9 nm) and C-atoms (156.1 nm) respectively. Sub-ppm sensitivity was achieved using line-of-sight absorption at these wavelengths.

An example data trace for C-atoms in shock-heated C_3O_2 - H_2 -Ar mixtures is shown in Fig. 12, along with an Arrhenius diagram for the rate coefficients inferred for $C + H_2 \rightarrow CH + H$. These rate data represent the first direct determinations of C-atom rate coefficients in high temperature gases, and they serve to illustrate the potential of the laser-photolysis shock tube in fundamental combustion kinetics studies.

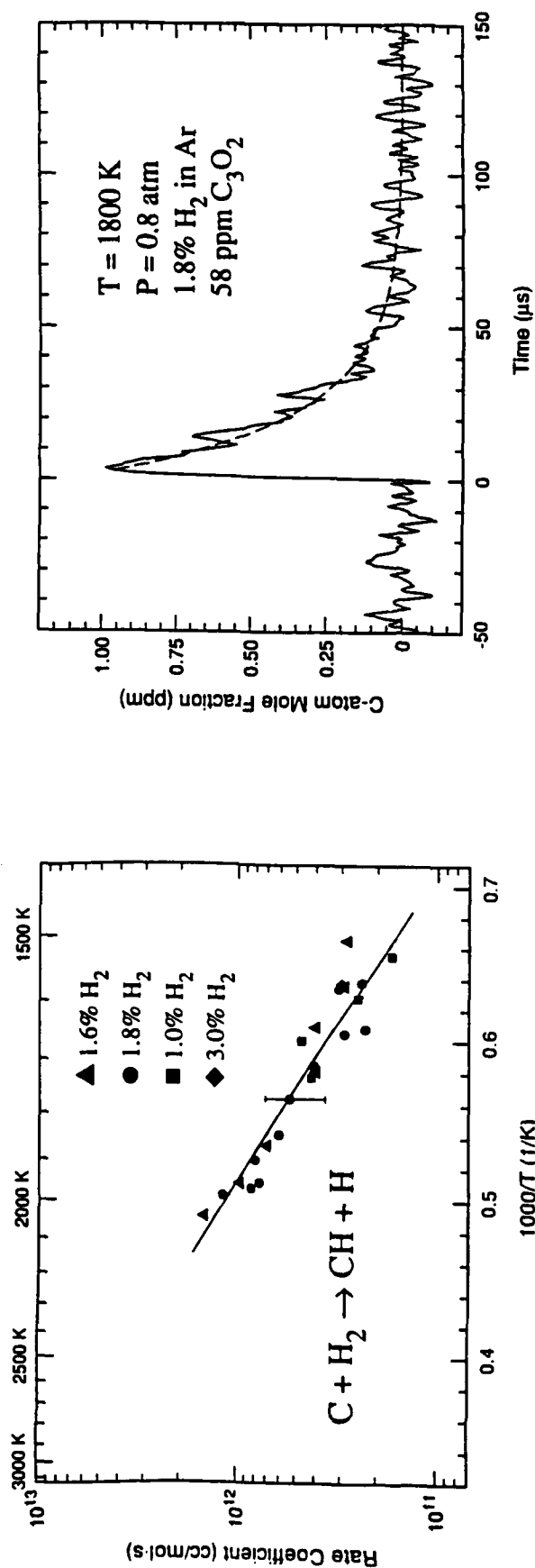
2.6 Digital Camera for High-Speed Imaging

The advancements made with PLIF as a diagnostic technique are tightly coupled to improvements in both laser sources and solid-state cameras. For this reason, a portion of our overall program is dedicated to advancing the state-of-the-art in lasers and solid-state camera systems. A major part of this effort in the past three years has been aimed at building a new camera with the capability for light-efficient, very-high-speed recording of PLIF images. This camera is the critical element needed to extend PLIF to: (1) "instantaneous" 3-d imaging; and (2) ultrafast 2-d imaging. The basic system, shown schematically in Fig. 13, is comprised of three critical elements: (1) a commercial image converter camera (Imacon 790 from Hadland); (2) a large (4 inch dia) tapered fiber-optic bundle; and (3) a large, high-resolution CCD array (400×1200 pixels from Reticon).

The image converter camera provides the basic fast-framing capability, allowing recording of up to 50 million frames per second (depending on the plug-in module) in a flexible output format containing a variable number of images (16 is a typical choice, though a larger number of lower-resolution images is possible). The output appears on a phosphor backplate and has traditionally been transferred to permanent storage with film. More recently, Hadland and others (e.g., a group from Yale and Sandia) have begun utilizing CCD arrays for recording the phosphor images. This has been done with lens coupling, however, which is much less efficient than transferring the images with a fiber bundle. Our approach to this problem has been to utilize a tapered fiber-optic bundle which matches (at one end) the output dimensions of the phosphor screen and (at the other end) the dimensions of the

LASER-PHOTOLYSIS SHOCK TUBE EXTENDED TO C- AND N-ATOM REACTIONS

- Direct Studies of Elementary Reactions Improve Understanding of Propellant Chemistry



- Method based on gasdynamic heating + laser (193 nm) photolysis
- Atom sources established for C-atom (C_3O_2) and N-atom (NO)
- First direct high temperature rate data for: $C + O_2 \rightarrow CO + O$



Figure 12. Extension of laser-photolysis shock tube to C- and N-atom reactions.

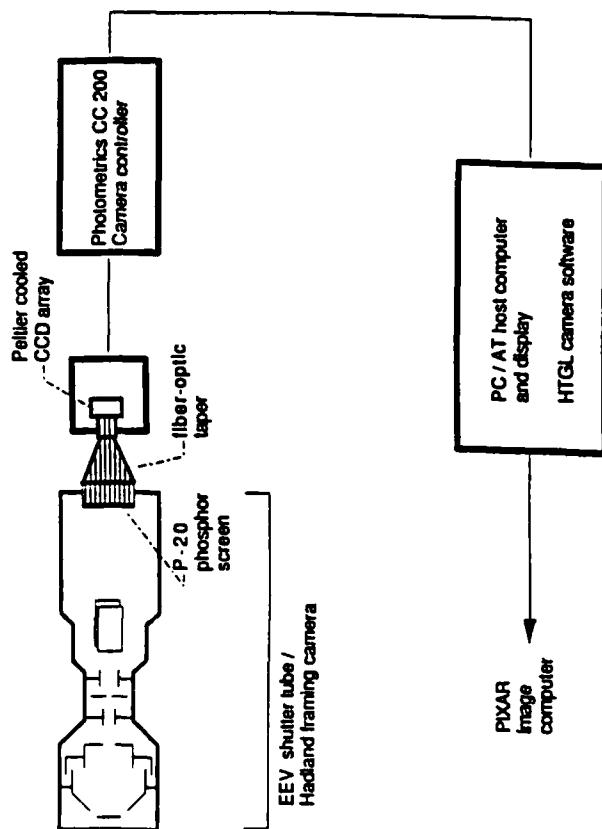
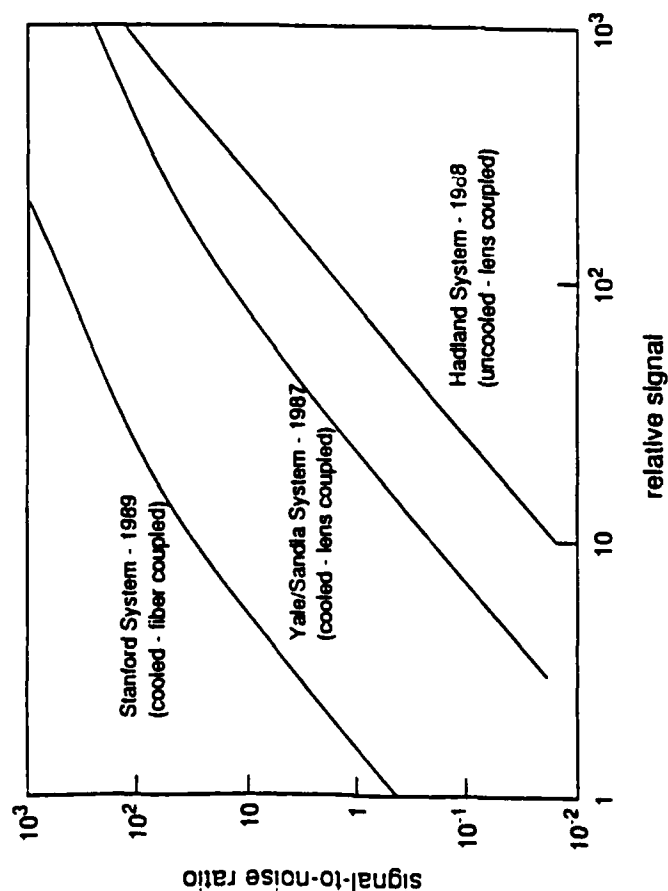
CCD array. This coupling scheme gives efficient transfer of light while maintaining high spatial resolution. The taper used is based on recently developed technology which allows production of bundles up to 4 inches in width. The last feature of the camera worth mention is the high-resolution, astronomy-grade CCD array which contains 480,000 pixels. This large format is nearly ideal for transferring the Imacon output to a CCD array without degradation of spatial resolution. Of course, once the images are stored on the array, the experimenter is free of the problems inherent with film and has access to all the advantages of on-line data processing and display.

During the past year, the last element of system, namely the CCD array and camera controller, were delivered and the overall camera was assembled at Stanford. It is worth noting that this is the first large-format CCD array fitted with a fiber optic stub, and it was necessary for the vendor (Photometrics, Inc.) to develop special grinding and test procedures to carry out this work. The camera is now undergoing bench-testing in our laboratory to measure critical performance parameters and to develop image correction procedures and algorithms to deal with the distortions introduced by both the fiber optic taper and the image converter camera. Although bench-testing is incomplete, our calculations of expected system performance, relative to previous image converter cameras, are indicated in Fig. 13. For the light levels expected in a representative PLIF imaging experiment, we estimate an improvement of about two orders of magnitude in signal-to-noise ratio relative to the current commercial camera system.

We have defined two types of experiments which will utilize the new camera: (1) 3-d PLIF imaging; and (2) 2-d time histories of PLIF images. In the first case, the goal is to be able to record an "instantaneous" 3-d image of the quantity being monitored by PLIF. This will be accomplished using a relatively long-pulse tunable dye laser (2 microsecond pulse length) which is formed into a light sheet and swept rapidly across the flow of interest. The camera will record a sequence of snap-shot PLIF images during this brief period, which taken together constitute a 3-d image with multiple 2-d planes. Such 3-d recording is important for several reasons, for example to provide direct measurements of 3-d flow structures and to allow examination of the directional dependence of properties such as dissipation. The second camera application, namely fast movies, is needed to allow study of very fast transients in a single 2-d plane. As an example, several plasma processes, such as laser ignition of combustion gases or electrical breakdown in electric propulsion devices, occur on very short time scales. These short times preclude use of current video cameras (which typically frame at up to 60 Hertz) and lasers (up to about 1 kilohertz) for time-resolving fast phenomena.

In summary, we expect this new camera to allow important extensions of PLIF (and other 2-d imaging schemes) into aerospace research areas where, at present, laser diagnostics have not yet been applied.

NEW DIGITAL CAMERA FOR HIGH-SPEED PLIF IMAGING



- System operates at 2k - 20M frames/sec.
- 10^2 x improvement in light throughput over commercial version.
- 10x improvement in spatial resolution.
- Use of CCD detection provides direct digitization of images.

Figure 13. New digital camera for high-speed PLIF imaging.

2.7 High-Resolution PLIF Imaging in a Turbulent Jet

One of the primary motivating issues for the development of planar laser-induced fluorescence imaging is the need for advanced diagnostics to study turbulence, and in particular turbulent combustion. The hope is that multi-dimensional data, of the type generated by PLIF, will provide improved insights into turbulence through the increased experimental information available. Since image data obtained are quite different from the data gathered with traditional single-point observations, new questions have arisen in identifying the experimental quantities of greatest value and in selecting the types of image processing to be performed in reducing image data. In order to address these diagnostics issues, we have worked over the past four years to perform imaging experiments in fundamental turbulent flowfields with the specific objectives of obtaining data with: (1) high signal-to-noise ratios; and (2) high spatial resolution. It was our belief that these two characteristics were necessary in order properly guide work on image processing and in order for the work to have significant impact on the fluid mechanics community. The flowfield selected for primary study was a turbulent, nonreacting round jet at atmospheric conditions. The jet was nitrogen, seeded with biacetyl to a mole fraction of about 5%, flowing into a pure nitrogen background. Excitation of the biacetyl fluorescence (and phosphorescence) was by a xenon fluoride excimer laser at 351 nm. This strategy satisfied the requirement for high SNR.

The high resolution imaging was done with a scientific grade unintensified CCD camera. This was the first application, to our knowledge, of the now popular 384x576 Thomson camera (currently available from Photometrics) to flowfield imaging. (It is important to note that the critical advantages inherent in imaging with an unintensified camera were enabled by both the high seeding levels allowed with biacetyl, owing to its high vapor pressure, and the fact that its emission is in the visible portion of the spectrum where the CCD array is sensitive.) Two sets of sample single-shot images obtained in these jets are shown in Fig. 14. The jet flows vertically downward from a 1 cm dia nozzle into a test chamber 60 cm x 60 cm in cross-section with a slow co-flow of nitrogen. The location of the various image planes is indicated on the figure, along with an indication of the size of the region imaged. The quantity being monitored (i.e., the quantity proportional to the scattered light intensity) is the jet fluid mole fraction, displayed here in a false color format. The important observation to be made is the very high SNR achieved, even though the images are based on molecular light-scattering phenomena. It is these high SNRs and the high spatial resolution obtained through use of a large CCD array which enable meaningful research on image processing of such data sets.

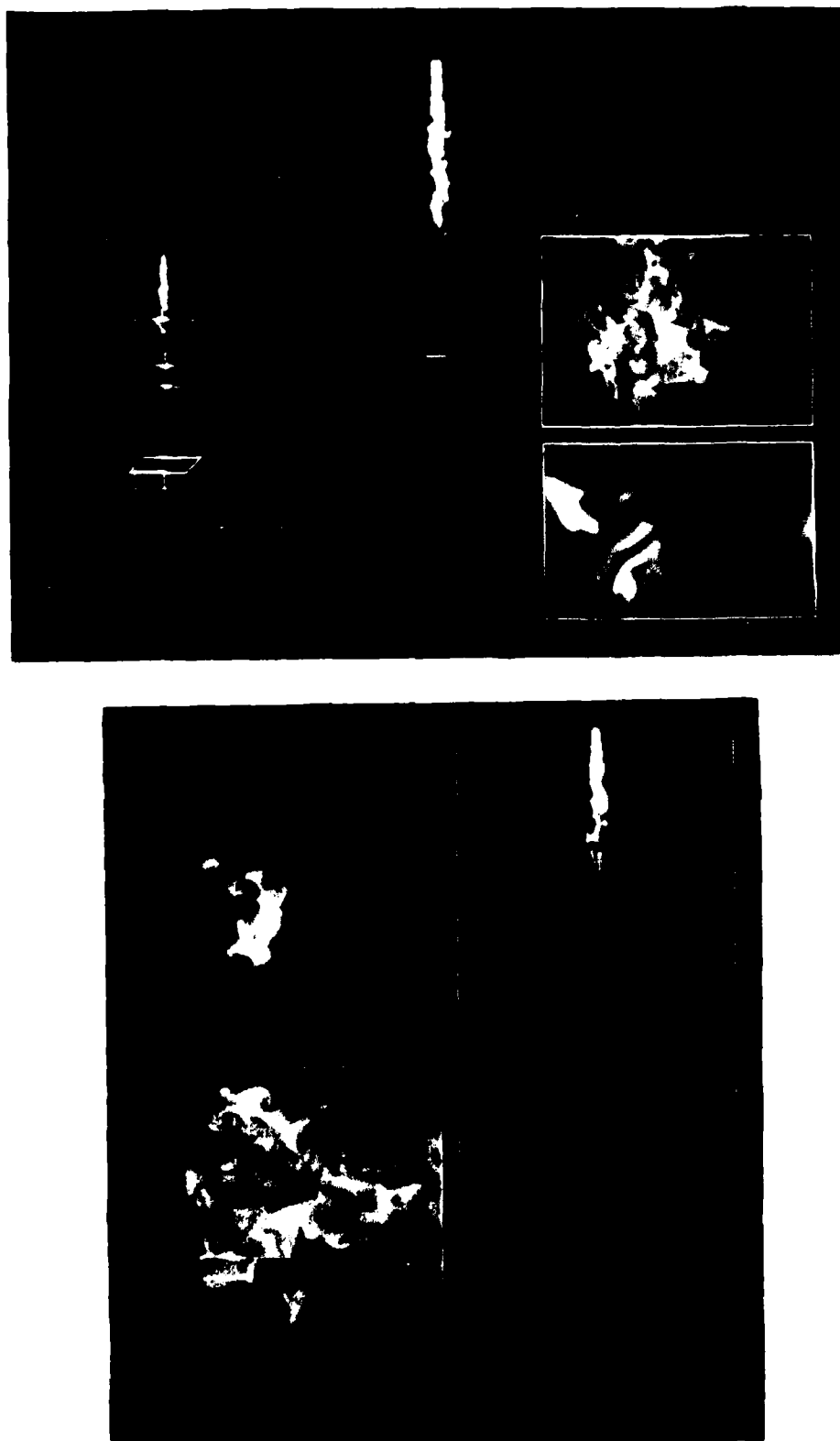


Figure 14. PLIF images in turbulent ($Re = 5000$) nonreacting N_2 jet seeded with biacetyl. Color coding of boxes indicates location and magnification of images.

Once recorded and transferred to permanent digital storage, the image data can be processed in several different ways, beginning with image corrections to allow for nonuniform illumination and light collection efficiencies. Simple examples of such processing are shown in Figs. 15 and 16. Figure 15 displays both corrected single-shot data and cross-sectional cuts through the data to illustrate radial, axial, and off-axis distributions. These data have been scaled by x (the distance downstream from the jet exit) to compensate for the declining signal level with axial distance. Note that the scaling restores the near-constant centerline signal level (consistent with the mean flow properties), and that the radial distributions are more nearly top-hat in form than gaussian (consistent with current theories/models of mixing).

Figure 16 illustrates the magnitude of "jet flapping," i.e., the motion of the jet fluid centroid. Panel (b) shows the average of 83 images of centroid locations, while panel (c) provides a plot of the distribution of 110 centroids at a fixed axial location 45 diameters downstream. The data in panel (c) are superposed on the mean cross-cut image. It is clear from the various plots of centroid position that jet-flapping plays a significant role in these flows and should be accounted for in mixing models. Note particularly the difficulty which would be encountered with single-point diagnostics in distinguishing between the effects of jet-flapping and the mixing occurring internal to the cross-section of jet fluid.

In summary, the focus of this project over the past year has been on the application end of PLIF rather than on technique development. Much of the research has been concerned with computer-based image processing of PLIF data, as we have sought to identify useful analysis schemes of these scalar data and useful correlations to guide turbulent-mixing modellers. An important result of this work will be an improved specification of measurement needs to guide the development of the next generation of imaging-based turbulent flow diagnostics. Example recommendations are likely to include: simultaneous measurement of two flowfield parameters, e.g. mixture fraction and velocity; measurements using multiple cameras for simultaneous recording of mixture fraction in different planes (adjacent or orthogonal); and double-pulse measurements for fixed planes to allow study of the temporal evolution of mixture fraction. Another likely recommendation will be experiments aimed at reacting jets, initially studied without heat release by employing a reactant (probably O_2) which highly quenches biacetyl phosphorescence; this has the same effect on the scattered light as a chemical reaction in "removing" a species.

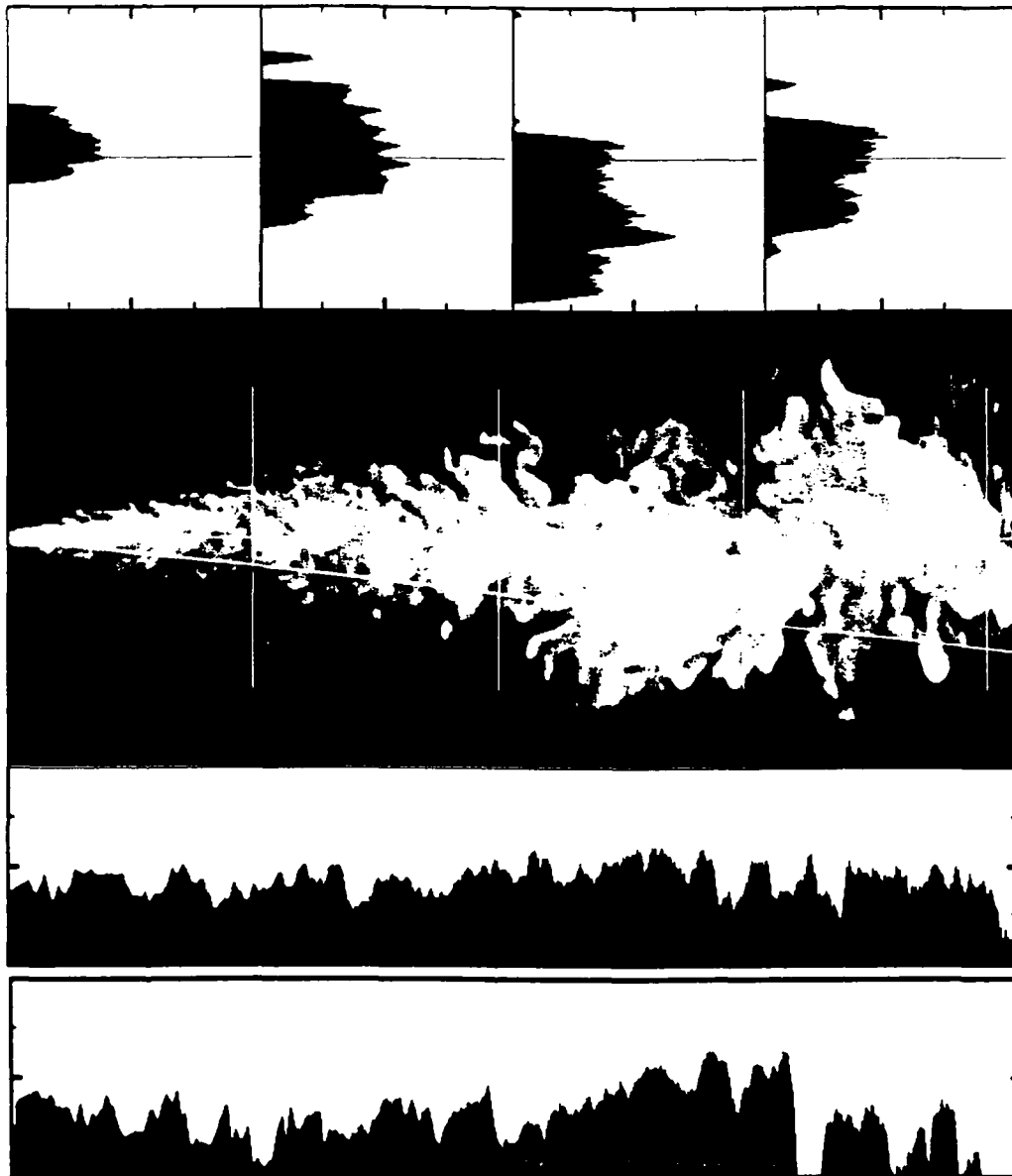


Figure 15. Instantaneous Concentration Field. 85 diameter field-of-view of $Re = 5000$ jet seeded with 5% mole fraction biacetyl. Cross-sections at 20, 40, 60, and 80 diameters displayed above the jet, and the centerline and 6 degree profiles displayed below the jet. The jet concentration has been self-similarly scaled using the scaling from the mean jet.

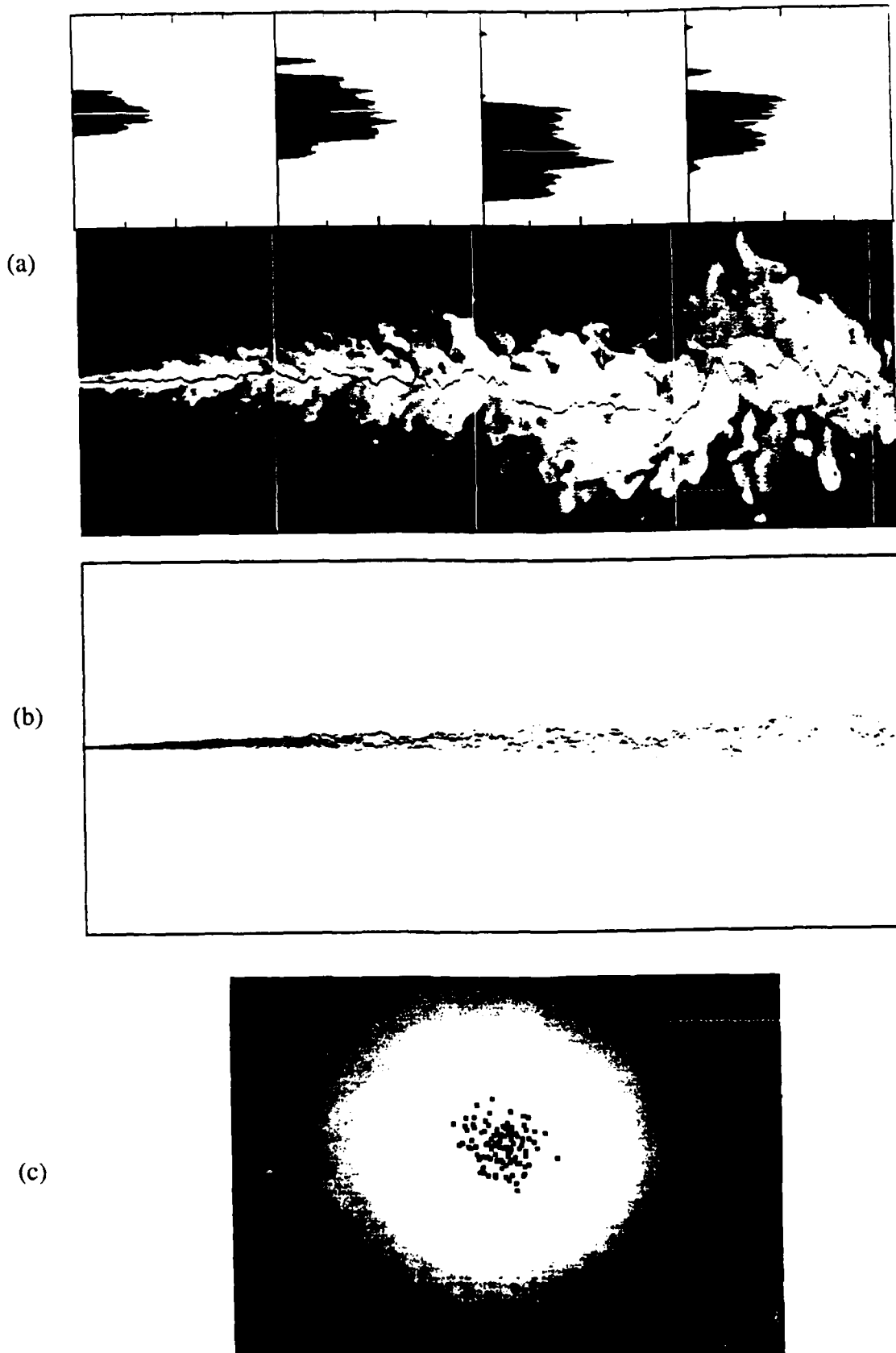


Figure 16. Cross-stream Concentration Centroid. (a) Centroid location superposed on an instantaneous jet image, (b) average of 83 images of centroid locaitons, and (c) 110 cross-cut centroids superposed on mean crosscut image from 45 diameters downstream.

3.0 PRESENTATIONS AND PUBLICATIONS

3.1 Presentations (10/89 – 10/90)

1. M. P. Lee, P. H. Paul, B. K. McMillin and R. K. Hanson, "Planar Laser-Induced Fluorescence Imaging of NO in Supersonic Flows," paper 89-40 at Fall WSS/CI Meeting, Livermore, CA, October 23-24, 1989.
2. J. M. Seitzman, A. Ungut, P. H. Paul and R. K. Hanson, "Structural Characterization of a Turbulent Nonpremixed Hydrogen-Air Flame with PLIF of OH," paper 89-42 at Fall WSS/CI Meeting, Livermore, CA, October 23-24, 1989.
3. E. C. Rea, Jr., A. Y. Chang and R. K. Hanson, "Motional Narrowing in Spectral Lines of OH," paper 89-45 at Fall WSS/CI Meeting, Livermore, CA, October 23-24, 1989.
4. D. F. Davidson, K. Kohse-Höinghaus, A. Y. Chang and R. K. Hanson, "A Pyrolysis Mechanism for Ammonia," paper 89-95 at Fall WSS/CI Meeting, Livermore, CA, October 23-24, 1989.
5. J. D. Mertens, A. Y. Chang, D. A. Masten, R. K. Hanson and C. T. Bowman, "A Shock Tube Study of the Reactions of NH with NO, O and O₂," paper 89-96 at Fall WSS/CI Meeting, Livermore, CA, October 23-24, 1989.
6. M. A. Cappelli, P. H. Paul and R. K. Hanson, "Laser-Induced Fluorescence Imaging of Laser Ablated Barium," presented at 42nd Gaseous Electronics Conf., Palo Alto, Oct. 1989.
7. D. F. Davidson, A. J. Dean, A. Y. Chang and R. K. Hanson, "Shock Tube Combustion Studies Using Optical Diagnostics and Excimer Photolysis," presented at 1989 AIChE Annual Meeting, San Francisco, CA, Nov. 5-10, 1989.
8. I. van Cruyningen, A. Lozano and R. K. Hanson, "Quantitative Planar Laser-Induced Fluorescence Imaging of Turbulent Jets," presented at 42nd annual meeting of the Fluid Dynamics Section of APS, NASA Ames Res. Center, CA, Nov. 19-21, 1989.
9. P. H. Paul, J. M. Seitzman, A. Ungut and R. K. Hanson, "Structural OH Imaging in a Turbulent H₂-Air Diffusion Flame," presented at 42nd annual meeting of the Fluid Dynamics Section of APS, NASA Ames Res. Center, CA, Nov. 19-21, 1989.
10. A. Lozano, I. van Cruyningen, M. G. Mungal and R. K. Hanson, "Volume Rendering of 2-D and 3-D Flowfield Image Data," presented at 42nd annual meeting of the Fluid Dynamics Section of APS, NASA Ames Res. Center, CA, Nov. 19-21, 1989.
11. M. P. Lee, P. H. Paul and R. K. Hanson, "2-D Velocity Measurements in Supersonic Flow Using Pulsed Planar Laser-Induced Fluorescence," presented at 1989 ASME Winter Meeting, Symp. of Flow Visualization, San Francisco, CA, December 10-15, 1989.

12. I. van Cruyningen, A. Lozano and R. K. Hanson, "Interpretation of Planar Laser-Induced Fluorescence Flowfield Images," presented at 1989 ASME Winter Meeting, Symp. of Flow Visualization, San Francisco, CA, December 10-15, 1989.
13. B. K. McMillin, M. P. Lee, J. L. Palmer, P. H. Paul and R. K. Hanson, "Planar Laser-Induced Fluorescence Imaging of Shock-Heated Flows in Vibrational Nonequilibrium," presented at 1989 ASME Winter Meeting, Symp. of Flow Visualization, San Francisco, CA, December 10-15, 1989.
14. J. M. Seitzman, R. K. Hanson, P. H. Paul, M. P. Lee and B. McMillin, "Laser-Induced Fluorescence Diagnostics for Supersonic Flows," paper WA2-1 at *Laser Applications to Chemical Analysis*, Feb. 5-8, 1990, Incline Village, NV.
15. R. K. Hanson, A. Y. Chang, J. M. Seitzman, M. P. Lee, P. H. Paul and B. E. Battles, "Laser-Induced Fluorescence Diagnostics for Supersonic Flows," reprint AIAA-90-0625 at AIAA 28th Aerospace Sciences Meeting, Reno, NV, Jan. 1990.
16. J. M. Seitzman, P. H. Paul and R. K. Hanson, "PLIF Imaging Analysis of OH Structures in a Turbulent Nonpremixed H_2 -Air Flame," reprint AIAA-90-0160 at AIAA 28th Aerospace Sciences Meeting, Reno, NV, Jan. 1990.
17. I. J. van Cruyningen, A. Lozano and R. K. Hanson, "Computer Rendering of Planar Fluorescence Flowfield Images," reprint AIAA-90-0499 at AIAA 28th Aerospace Sciences Meeting, Reno, NV, Jan. 1990.
18. R. K. Hanson, "Planar Fluorescence Imaging: Concepts and Applications," invited presentation at NATO Advanced Study Institute, Algarve, Portugal, April 16-27, 1990.
19. B. K. McMillin, M. P. Lee, P. H. Paul and R. K. Hanson, "Planar Laser-Induced Fluorescence Imaging of Shock-Induced Ignition," presented at Twenty-Third Symposium (International) on Combustion, Orleans, France, July, 1990.
20. D. F. Davidson and R. K. Hanson, "Shock Tube Measurements of the Rate Coefficient for $N + CH_3 \rightarrow H_2CN + H$ using N-Atom ARAS and Excimer Photolysis of NO ," presented at Twenty-Third Symposium (International) on Combustion, Orleans, France, July, 1990.
21. J. M. Seitzman, A. Ungut, P. H. Paul and R. K. Hanson, "Imaging and Characterization of OH Structures in a Turbulent Nonpremixed Flame," presented at Twenty-Third Symposium (International) on Combustion, Orleans, France, July, 1990.
22. P. H. Paul, M. P. Lee, B. K. McMillin, J. M. Seitzman and R. K. Hanson, "Application of Planar Laser-Induced Fluorescence Imaging Diagnostics to Supersonic Reacting Flow," paper 90-1844 at 28th AIAA/SAE/ASME/ASEE Joint Propulsion Conf., Orlando, FL, July, 1990.
23. R. K. Hanson, A. Y. Chang, M. D. DiRosa, L. C. Philippe, B. K. McMillin and M. P. Lee, "Laser-Based Diagnostics for Propulsion and Hypersonics Testing," paper AIAA-90-1383 presented at AIAA 16th Aerodynamic Ground Testing Conf., Seattle WA, June 1990.

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3.2 Publications (10/89 – 10/90)

1. B. Hiller, P. H. Paul and R. K. Hanson, "Image-Intensified Photodiode Array as a Fluorescence Detector in CW-Laser Experiments," Review of Scientific Inst., **61** (7), 1808-1815 (1990).
2. B. Hiller and R. K. Hanson, "Properties of the Iodine Molecule Relevant to Absorption/Fluorescence Experiments in Gas Flows" Experiments in Fluids, in press.
3. A. J. Dean and R. K. Hanson, "Development of a Laser Absorption Diagnostic for Shock Tube Studies of CH," J. Quant. Spectrosc. and Radiat. Trans. **42**, 375-384 (1989).
4. P. H. Paul, M. P. Lee, B. K. McMillin, L. M. Cohen and R. K. Hanson, "Planar Laser-Induced Fluorescence Imaging in Supersonic Flows," AIAA Journal, in press.
5. A. J. Dean, D. F. Davidson and R. K. Hanson, "C-Atom ARAS Diagnostic for Shock Tube Kinetics Studies," AIP Conf. Proc. **208**, *Current Topics in Shock Waves: 17th Shock Tube Symp.*, ed. Y. W. Kim (Bethlehem, PA 1990), pp. 537-542.
6. D. F. Davidson, D. C. Snell and R. K. Hanson, "Shock Tube Excimer Photolysis and the Measurement of N Atom Kinetic Rates," AIP Conf. Proc. **208**, *Current Topics in Shock Waves: 17th Shock Tube Symp.*, ed. Y. W. Kim (Bethlehem, PA 1990), pp. 5257-530.
7. D. F. Davidson, K. Kohse-Höinghaus, A. Y. Chang and R. K. Hanson, "A Pyrolysis Mechanism for Ammonia," Int. J. of Chem. Kinetics **22**, 513-535 (1990).
8. J. D. Mertens, A. Y. Chang, R. K. Hanson and C. T. Bowman, "Reaction Kinetics of NH in the Shock Tube Pyrolysis of HNCO," Int. J. of Chem. Kinetics **21**, 1049-1067 (1989).
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10. M. P. Lee, P. H. Paul and R. K. Hanson, "2-D Velocity Measurements in Supersonic Flow Using Pulsed Planar Laser-Induced Fluorescence," ASME FED-Vol. 85, pp. 101-108 (1989).
11. I. van Cruyningen, A. Lozano and R. K. Hanson, "Interpretation of Planar Laser-Induced Fluorescence Flowfield Images," ASME FED-Vol. 85, pp. 109-114 (1989).

12. A. Y. Chang, B. E. Battles and R. K. Hanson, "Simultaneous Measurements of Velocity, Temperature and Pressure using Rapid cw Wavelength-Modulation LIF of OH," *Optics Letters* **15**, 706-708 (1990).
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14. I. van Cruyningen, A. Lozano, M. G. Mungal, and R. K. Hanson, "3D Visualization of Temporal Flow Sequences," *AIAA Journal*, in press.
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16. P. H. Paul, I. van Cruyningen, R. K. Hanson and G. Kychakoff, "High Resolution Digital Flowfield Imaging of Jets," *Experiments in Fluids* **9**, 241-251 (1990).
17. I. J. van Cruyningen, A. Lozano and R. K. Hanson, "Computer Rendering of Planar Fluorescence Flowfield Images," reprint AIAA-90-0499 at AIAA 28th Aerospace Sciences Meeting, Reno, NV, Jan. 1990.
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20. J. M. Seitzman, A. Ungut, P. H. Paul and R. K. Hanson, "Imaging and Characterization of OH Structures in a Turbulent Nonpremixed Flame," *Twenty-Third Symposium (International) on Combustion*, The Combustion Institute, in press.
21. I. van Cruyningen, A. Lozano and R. K. Hanson, "Quantitative Planar Laser-Induced Fluorescence Imaging of Concentration," *Experiments in Fluids*, in press.
22. P. H. Paul, M. P. Lee, B. K. McMillin, J. M. Seitzman and R. K. Hanson, "Application of Planar Laser-Induced Fluorescence Imaging Diagnostics to Supersonic Reacting Flow," paper 90-1844 at 28th AIAA/SAE/ASME/ASEE Joint Propulsion Conf., Orlando, FL, July, 1990.
23. A. J. Dean, R. K. Hanson and C. T. Bowman, "A Shock Tube Study of Reactions of C-atoms and CH with NO including Product Channel Measurements," *J. Phys. Chem.*, in press.

4.0 PERSONNEL

Individual researchers supported by the program are listed below. All the work has been carried out in the High Temperature Gasdynamic Laboratory, in the Department of Mechanical Engineering, under the supervision of Professor R. K. Hanson.

Research Associates

Dr. P. H. Paul

Dr. D. F. Davidson

Graduate Research Assistants

Jerry Seitzman

Mike Lee

Ike van Cruyningen

Doug Baer

Brian McMillin

Dave Hofeldt

5.0 SIGNIFICANT INTERACTIONS

In addition to the interactions associated with the presentations and publications listed in Section 3, we have had numerous visitors to our laboratory during this past year. Foreign visitors have come from Germany, France, Holland, Great Britain, Canada, Spain and Japan; industrial and national laboratory visitors have included representatives from Rocketdyne, Physical Sciences, Lockheed, Boeing, Metrolaser, AEDC, NASA Ames, NASA Lewis, NIST, Sandia, Lawrence Livermore, General Motors and Ford. Professor Hanson has given invited presentations on AFOSR-sponsored diagnostics research to industrial laboratories and government groups. Members of our group have provided technical information and advice, by telephone and mail, to several external researchers interested in duplicating or extending our diagnostics concepts.

Interest in the potential application of advanced laser diagnostics to various practical problems, especially associated with hypersonic flow and the NASP program, continues to grow, and the AFOSR-sponsored program at Stanford has achieved a high level of recognition for its contributions to this field. The increased interest we are witnessing is the leading edge of technology transfer of laser diagnostics to industrial and government labs. A sustained research effort, at Stanford and other university labs active in diagnostics research, is required, however, to ensure the success of this technology transfer.